

Molecular transitions in Fermi condensates

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We discuss the transition of fermion systems to a condensate of Bose dimers, when the interaction is varied by use of a Feshbach resonance. We argue that there is an intermediate phase between the superfluid Fermi gas and the Bose condensate of molecules, consisting of extended dimers.

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The remarkable experimental advances in atomic trap physics now permits one to study Fermi gases of atoms over a broad range of couplings from weak to strong. In particular, two-component Fermi systems have been made and studied using the two alkali metal isotopes ⁴⁰K [1, 2, 3] and ⁶Li [4, 5, 6]. In both cases, the Feshbach resonance phenomenon has been exploited to vary the coupling strength of the interaction between atoms and study the consequent many-particle properties. In both cases there is a closed channel near threshold whose energy can be tuned with respect to the open two-body channels. A particularly interesting feature is the formation of molecules or dimers as the effective interaction strength is increased. There are many questions that can be posed about a coexisting phase of bound pairs and that we would like a theory to address.

Here we want to argue that the theoretical situation is rendered more complex by the fact that there are in fact two kinds of pair states that must be treated in any theory that applies to the full range of couplings. These two kinds of pairs are spatially different and have different quantum numbers as well. The first bound state appears when the inverse scattering length passes through zero (via the tuning of the Feshbach resonance). We shall call it the *halo dimer*, in analogy with somewhat similar *halo nuclei* [7]. A halo dimer is spatially extended and has only a small overlap with the closed-channel Feshbach resonance. But as the closed-channel resonance is tuned to lower energies, its energy eventually becomes negative and can be identified with the pair state. We present below a simple model that shows how this two-step transition from the continuum pair states to the molecular bound state takes place, deriving the relevant size and energy scales. We then discuss the experimental signatures of the halo dimer state. Finally, we assess the theoretical approaches that have been applied to treat the many-particle system in the halo dimer domain. There are a number of calculations in the literature that ignore the special character of these states, and we believe that such models lead to a number of falsifiable predictions.

We consider here a simple model of the two-body physics using a two-channel atomic Hamiltonian of the form

$$\begin{bmatrix} H_{11}(r) & V_{12}(r) \\ V_{21}(r) & H_{22}(r) \end{bmatrix} \begin{bmatrix} u(r) \\ v(r) \end{bmatrix} = k^2 \begin{bmatrix} u(r) \\ v(r) \end{bmatrix}, \quad (1)$$

where r is the atom-atom separation. We shall use the units $\hbar = m = 1$ and here $u(r)$ describes the open two-atom channel and $v(r)$ the closed (molecular) channel. It is implied that while $V_{11}(r) \rightarrow 0$ and $V_{12}(r) = V_{21}(r) \rightarrow 0$ when $r \rightarrow \infty$, the closed channel potential tends to a positive constant $V_{22}(r) \rightarrow U_0 > 0$. Since we are interested in very low energies, only the s -wave is considered here and we show only the radial parts of the wave functions. If one were to apply this model to a system of ⁶Li atoms, in the open channel at magnetic fields near the Feshbach resonance the two valence electrons of the two ⁶Li atoms would be anti-parallel to the magnetic field (triplet state), while in the closed channel the two electron spins would be anti-parallel to each other (singlet state). As in all previous references [8, 9, 10, 11, 12] we shall assume that in the closed channel there is only one state close to the two-atom threshold. If the wave function of this state is $\phi_0(r)$ with an energy κ_0^2 with respect to the two-atom threshold, one can easily show that the two-channel problem simplifies somewhat and the wave functions read

$$u(r) = u_0(r) + \left\langle r \left| \frac{1}{k^2 - H_{11}} V_{12} \right| \phi_0 \right\rangle \frac{\langle \phi_0 | V_{21} | u \rangle}{k^2 - \kappa_0^2}, \quad (2)$$

$$v(r) = \phi_0(r) \frac{\langle \phi_0 | V_{21} | u \rangle}{k^2 - \kappa_0^2}, \quad (3)$$

$$H_{11}(r)u_0(r) = k^2 u_0(r), \quad (4)$$

$$H_{22}(r)\phi_0(r) = \kappa_0^2 \phi_0(r), \quad (5)$$

For the sake of simplicity, we shall assume that in the open channel the atoms are free (the same assumption was made in Ref. [8]), that $\phi_0(r) = \sin(\pi r/r_0)\sqrt{2/r_0}$ for $0 \leq r \leq r_0$ and zero otherwise and that $V_{12}(r) = V_{21}(r) = g\delta(r - r_1)$, with $r_1 = r_0/2$. Physically, the parameter r_0 is of the order of the van der Waals length $r_0 \approx (C_6 m/\hbar^2)^{1/4}$ and we shall consider only such energies for which $kr_0 \ll 1$. After some simple manipulations one arrives at the following form of the two-atom wave

function in the open channel (using $u_0(r) = \sin(kr)$)

$$u(r) = \sin(kr) + \frac{g^2 \exp(ikr_>) \sin(kr_<)}{\kappa_0^2 - g^2 - ig^2 kr_1}, \quad (6)$$

where $r_> = \max(r, r_1)$ and $r_< = \min(r, r_1)$ and terms of order $\mathcal{O}(k^2)$ have been neglected. One can then easily show that

$$-\frac{1}{a} = \frac{\kappa_0^2 - g^2}{g^2 r_1}, \quad (7)$$

$$u(r) = \exp(i\delta) \sin k(r - a), \quad \text{if } r \geq r_1, \quad (8)$$

where $\tan \delta = -ka$. In particular, exactly at the resonance $u(r) = i \cos(kr)$ for $r \geq r_1$. Far away from the Feshbach resonance, the open channel wave function would be approximately equal to $u(r) = \sin(kr)$ instead (for $r \geq r_0$). By the nature of the problem at hand we have $\kappa_0 r_0 = \mathcal{O}(1)$ and $gr_0 = \mathcal{O}(1)$. Only if κ_0 and g are comparable in magnitude one can attain the regime when $a = \pm\infty$. For the experiments reported, the relative energy of the Feshbach resonance and thus the magnitude of κ_0^2 is controlled by a magnetic field. With fine tuning one can make the numerator in Eq. (7) very small, and in this way attain the regime $|a| \gg r_0$, even though all the parameters in the equation above are of order $1/r_0$ or r_0 respectively. As an order of magnitude estimate for these constants one can use $\mathcal{O}(\kappa_0^2) = \mathcal{O}(g^2) = 2\mu_B B_0 m / \hbar^2$, where m is the atomic mass, μ_B is Bohr magneton and B_0 is the value of the magnetic field where $1/a = 0$. For both ^6Li and ^{40}K one readily obtains that $\mathcal{O}(\kappa_0 r_0) = \mathcal{O}(gr_0) = 1$.

Because of the coupling between the two channels, there is now a pole of the scattering amplitude at $k_0 = i/a$, as may be seen from Eq. (6). In different terms, by means of a magnetic field one controls the logarithmic derivative of the open channel wave function near the origin, more exactly near $r = r_0$. In a finite density medium it might naively appear that an infinite value of the scattering length would be meaningless. However, that should be interpreted rather as the logarithmic derivative of the open channel wave function at $r = r_0$, namely $d \ln u(r)/dr|_{r=r_0} = -1/a$, or in more physical terms, as the relative momentum with which the two atoms emerge after interacting at short distances. When the two-atom system is far from the Feshbach resonance the typical relative momentum with which the two atoms emerge after interacting at distances smaller than r_0 is \hbar/r_0 . The special situation, which is achieved by bringing the two atoms exactly at the Feshbach resonance, is to insure that they emerge from the interaction region with an essentially vanishing relative momentum. In a certain sense that amounts to an ultimate further cooling of the relative atomic motion to its minimum and that is what makes the physics of atoms under these conditions particularly exciting. In a sense, two-atom collisions at short distances do not bring in any momentum.

TABLE I: Character of the condensate as a function of the inverse scattering length a^{-1} in various intervals, the approximate boundaries of these intervals being shown in the second row. The total electron spin and spin projection (S, S_z) along the magnetic field for various pairs are shown in the last row.

$a^{-1} > 0$			$a^{-1} < 0$	
$+\infty$	r_0^{-1}	k_F	0	$-\infty$
molecules	halo dimers (+ atoms ?[15])	?	BCS strong coupling	BCS weak coupling
(0,0)	(1,-1)	(1,-1)	(1,-1)	(1,-1)

Let us put this two-atom system in a spherical cavity of radius $R \gg r_0$. In principle one would have to specify the boundary condition for $u(r)$ at $r = R$, which would lead to energy quantization. The specific nature of this energy quantization (e.g. Neumann *vs* Dirichlet boundary conditions) is qualitatively unimportant. By choosing the radius of this spherical cavity one can simulate a Fermi gas of various densities, with $k_F \propto 1/R$, and in particular choose the regime with $k_F |a| \gg 1$, when according to the authors of Ref. [8] the two atoms should be with probability essentially one in the small size molecular state. One has simply to estimate the ratio of the probability to find the atoms in one or another channel, and one easily finds that

$$\int_0^R dr |v(r)|^2 = \frac{k^2 a^2}{(k^2 a^2 + 1) g^2 r_1} \leq \frac{1}{g^2 r_1} = \mathcal{O}(r_0), \quad (9)$$

$$\int_0^R dr |u(r)|^2 \approx \int_{r_0}^R dr \sin^2(kr + \delta) = \mathcal{O}(R). \quad (10)$$

Consequently, the relative probability to find the two atoms in the molecular state is of the order $\mathcal{O}(r_0/R) = \mathcal{O}(k_F r_0) \ll 1$ for a dilute Fermi gas, $nr_0^3 \ll 1$.

One can continue the argument in the region of negative detuning, when $a > 0$, and easily convince oneself that as long as $a \gg r_0$, the relative probability to find the two atoms in the closed channel is small. That was discussed in Ref. [13] and shown in another explicit calculation in Ref. [14]. Only when the scattering amplitude becomes of the order of the radius of the interaction (van der Waals length) the probability to find two atoms in the closed channel becomes comparable with the probability to find the same atoms in the open channel. The various regimes of the coupling strength are shown in Table I.

Next we discuss experimental observables to distinguish between the two kinds of bound states. *i)* One signature is magnetic. The experiments are typically carried out in magnetic fields large compared to the hyperfine splitting. The continuum states are thus polarized with respect to the electron spin, with the nuclear spin mainly responsible for the second component of a two-component Fermi gas. Thus the electron spin wave function of the halo dimer is largely $S=1$. The closed

channel, on the other hand, is typically well described with a spin-singlet electron wave function. In the experiment of Ref. [5], the spin of the bound pair was measured (see the insert of Fig. 2), and it was indeed found that its value was large down to values of the magnetic field well below the value at which the scattering length changes sign. This result is in perfect agreement with microscopic understanding of the Feshbach resonance in ^6Li [16]. *ii)* Another signal of the character of the pair state in the condensate is in the particle loss rate of the system. When the condensate is prepared with the atoms in the lowest hyperfine states, the only inelastic processes are three-body collisions in which one of the pairs goes into a deeply bound molecule. Such processes are more probable when three particles are simultaneously in each other's range of interaction. The observations of Ref. [5], see Fig. 4, confirmed by a separate experiment Ref. [3], in fact show that the loss rate becomes large under the same conditions that the bound pair develops a singlet character. This observation also agrees with the theoretical expectation of Ref. [17]. *iii)* We mention one more piece of evidence, this one more indirect. As several experimental groups have demonstrated [6] when the scattering length is positive (thus on the BEC side of the Feshbach resonance) and when $na^3 < 1$ the sizes of the atomic clouds agree very well with theory based on halo dimers, which predicts that the interaction between them can be characterized by a scattering length of magnitude $0.6a$ [15, 17]. If the pairs were in closed-channel molecular state, their scattering length would have the order of magnitude of their size $\mathcal{O}(r_0)$ independent of the atom-atom scattering length a .

We now turn to theoretical models for the condensation of trapped fermions into molecules. There have been a number of recent calculations [8, 9, 10, 11], based on the model of Timmermans *et al.*, [12] that assume a direct transition between the fermions and a molecular bound state. In Ref. [8] it was claimed that a dilute atomic Fermi gas near a Feshbach resonance undergoes a crossover into diatomic molecules of relatively small sizes on the BCS side of the resonance, when the atomic scattering length is still negative and the molecular energy level lies in the two-atom continuum. The authors of Ref. [8] describe the results of the recent experiment of Regal *et al.* [2], by the way of this conversion of atoms into diatomic molecules. Another recent preprint makes a similar claim, that exactly this process occurs on the BCS side of the Feshbach resonance [9], although this particular work deals with different properties of such systems.

The statement we take issue with here was formulated extremely succinctly by the authors of Ref. [8]. One considers a uniform dilute Fermi gas of number density $n = k_F^3/3\pi^2$, near a Feshbach resonance at positive detuning ($a < 0$), when the energy of the molecular state is $\epsilon_m \simeq \hbar^2/ma^2 > 0$. With respect to the two-atom contin-

uum this molecular state is unbound. Then if $k_F|a| > 1$ a fraction of these atoms converts into molecules with a number density, see Ref. [8] and also Refs. [9, 11] where a similar result is quoted,

$$n_{mc} \simeq \frac{n}{2} \left[1 - \frac{1}{(k_F|a|)^3} \right]. \quad (11)$$

Naively this results seems obvious, since as soon as the Fermi energy exceeds the (positive) molecular state energy, the system can only lower its total energy by converting into molecules. These molecules, being bosons in character, can all have zero center of mass kinetic energy, and thus the Fermi energy is lowered until it becomes equal to $\epsilon_m \simeq \hbar^2/ma^2$ and further conversion of atoms into molecules becomes energetically impossible. Applying this to the two-body problem, the transition would take place discontinuously as the inverse scattering length approaches zero from the BCS side. But as we saw previously, the formation of halo dimers is a smooth process with the two-particle wave function changing smoothly as the inverse scattering length goes through zero. It is also abundantly clear that such models [8, 9, 10, 11] would predict the wrong magnitude for the magnetic moments of such Fermi clouds near the Feshbach resonance. If indeed the BEC of small size molecules would occur in this regime, the prediction would be that the magnetic moment of such pairs would be vanishing, while experiment clearly shows that not to be the case [5].

There is still one possibility, when the mechanism suggested in Refs. [8, 9, 10, 11] could prove indeed correct and near a Feshbach resonance such a system would condense into molecules of relatively small size $\mathcal{O}(r_0)$. This situation would occur if the coupling constant g becomes unnaturally small. The relative probability that two atoms spend most of the time in a small size configuration, according to Eqs. (9 - 10) would be $\propto 1/(g^2 r_1 R) \approx k_F r_0 / (g r_0)^2$, which could become accidentally large, even though the system is still dilute, in the sense that $k_F r_0 \ll 1$. This would happen if the Feshbach resonance would be accidentally extremely close to the atom-atom threshold. Whether this mechanism can be indeed realized in experiments with dilute atomic Fermi clouds, is a question that remains to be investigated.

Since the halo dimers occupy the same sector of the two-particle Hilbert space as the plane waves of the Fermi gas, it is reasonable to ask whether a theory can be found that does not introduce a closed channel resonance explicitly. In fact, the BCS theory has the degrees of freedom to describe the many-particle system over the full range of couplings we consider here [18]. One may show [19] that the BCS theory contains the correct description of the gas of halo dimers in the limit that $a \ll 1/k_F$ and positive. Namely, the BCS chemical potential μ is

related to the dimer energy E_d by

$$\mu = \frac{E_d}{2} = -\frac{\hbar^2}{2ma^2}. \quad (12)$$

On the other hand, the BCS theory is quantitatively incorrect on at least two properties. The pairing gap Δ is reduced by polarization effects from the BCS value by about a factor of ≈ 2 both in the weak [20] and strong coupling limit [21]. Also, the dimer-dimer scattering amplitude is predicted to be $2a$ in the BCS theory [22], while a more detailed treatment gives $0.6a$ [15, 17]. Still, it is still possible to use the a BCS framework for an effective theory, renormalizing the couplings to reproduce these interactions, in the framework of the Superfluid LDA (SLDA) [23].

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